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Mathematical and computational approaches for design of biomass gasification for hydrogen production: A review

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ABSTRACT

The ever growing environmental concern caused by excessive use of fossil fuels in energy and transportation systems triggered considerable investigations on alternative energy sources such as biomass. Furthermore, the availability and security of fossil fuels to meet future global energy need are also subjected to uncertainty. For these reasons, the world's current focus is shifted towards hydrogen-based future economy. Gasification is a proven technology to produce satisfactory yield of hydrogen. Many studies have been performed to increase the production yield. Due to the extensive range of investigations, mathematical and computational approaches have been applied to conduct these studies. Thus, this paper aims to update and broaden the review coverage by incorporating works done to materialize the investigations on the potential of producing hydrogen from biomass via gasification encompassing mathematical modeling, simulation, optimization, process heat integration and cogeneration. Each of these subjects is reviewed and analyzed which helped to identify their respective strength and areas which require further research effort.

Contents

Abbreviations: BFBG, bubbling fluidized bed gasifier; CFBG, circulating fluidized bed gasifier; TBFBG, twin-bed fluidized bed gasifier; D, dimension.

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1. Introduction

In recent years, renewable types of energy have received much attention worldwide. Due to its advantageous characteristics, hydrogen is expected to satisfy a considerable, if not a major, portion of world's future energy need as enlightened by a number of researchers [1–6]. Once produced—water being its only combustion product—hydrogen has started to gain ground as an eco-friendly fuel [7,8]. Moreover, it has by far the highest energy content, i.e. 141.8 MJ/kg, in comparison to other common fuels [9]. Although fossil fuels i.e. natural gas, heavy oil, naphtha and coal make up the largest current source of hydrogen (i.e. ~97% [10,11]), their depletion concerns along with high carbon footprint [11–14] leads to the immense investigation on renewable alternative source of hydrogen [15]. Water electrolysis is also reported to be energy intensive resulting in high hydrogen production cost, e.g. 4.36–7.36 $/kg [16]. In contrast, some exclusive features of biomass including local availability, large commercialization potential, carbon-neutral nature and being a renewable energy resource, make the route feasible for sustainable hydrogen production [17–20].

Hydrogen fuel has a wider range of applications including domestic, industrial and space uses [21,22]. In this paper, the use of hydrogen in combined heat and power (CHP) generation is of particular discussion interest (Section 6) since the technology ensures high system efficiency and supports the integration of the sustainable biomass-based hydrogen into the existing large energy systems [23].

Biomass gasification is a proven technology to produce satisfactory yield of hydrogen. Many studies have been performed to increase the production yield. Due to the extensive range of investigations, mathematical and computational approaches have been applied to conduct these studies. This paper aims to present updated review on mathematical and computational works that are closely contributing to the development of hydrogen production technology via gasification of biomass. The overview of the technology is given in Section 2. The mathematical modeling works analyzed are categorized into equilibrium, kinetic and neural networks modeling approaches. Review on existing software based simulation models, optimization, heat integration and cogeneration works is also included (Sections 3–7). The areas which need further research and development effort are also mentioned.

2. Biomass conversion into hydrogen

As biomass is characterized by a low energy density, converting its energy content into more practical and clean gaseous fuel, i.e. hydrogen represents a successful option [24]. Based on the type and physical state of biomass, thermo-chemical conversion (conventional combustion, pyrolysis and gasification), bio-chemical conversion (fermentation and anaerobic digestion) and mechanical extraction are technologies applied for converting biomass into hydrogen [25]. Kalinci et al. [26] reviewed the literatures on the various methods exploited for biomass based hydrogen production. Biagini et al. [24] conducted experimental study to evaluate the performance of the different thermo-chemical configurations (i.e. combustion, gasification, electrolysis and syngas separation) for hydrogen production from biomass. The authors reported that the hydrogen production was maximized for the gasification/separation process followed by the gasification/electrolysis and the least being the combustion/electrolysis. With respect to related researches in Malaysia, Ahmad et al. [27] has recently reported the potential development of producing hydrogen from biomass.

2.1. Gasification of biomass into hydrogen

On the whole, among the aforementioned technology options, gasification of biomass is identified as the most efficient and economical route for hydrogen production [4,28]. Gasification is a high-temperature partial oxidation process in which a solid carbonaceous feedstocks such as biomass is converted into a gaseous mixture (H_2, CO, CO_2, CH_4, light hydrocarbons, tar, char, ash and minor contaminates) using gasifying agents [29,30]. The primary emphasis in biomass gasification is to maximize the yield of the product gas which in turn ensures high hydrogen yield. The performance of biomass gasification processes is influenced by at least 20 operation parameters concerning the gasifier and feedstock such as flow rate, composition and moisture content of the feedstock, geometrical configuration of the gasifier, reaction/residence time, type of gasifying agent, gasification temperature and pressure, the gasifying agent/biomass ratios, etc. [31,32].

Nevertheless, the development of hydrogen production from biomass gasification is still facing several challenges [33]. For instance the presence of tar which is an undesirable condensable organic compound in gasification product gas restricts the quick and widespread use of hydrogen. Unless the tar content in the product gas is controlled it imposes various problems including fouling and plugging in end-use devices such as exit pipes, heat exchangers, particulate filters and gas turbines and difficulties in handling tar–water mixtures and contamination of waste streams [34]. Moreover, since hydrogen has low energy content by volume its storage mechanism also requires additional research effort for its effective and efficient implementation in areas where size and weight constraints are dominant.

2.2. Gasifying agents

Air, pure oxygen, steam, carbon dioxide [35], nitrogen [36] or mixtures are the known gasifying agents used in biomass gasification [30]. The different gasifying agents produce gas with different calorific value. Even though air gasification offers operation simplicity and lack of dependence on complex industrial infrastructures and utilities [37], the technology produces low heating value gas, i.e. 4–7 MJ/Nm^3 [38]. Conversely, oxygen and steam gasification produce medium heating value gas, i.e. 10–18 MJ/Nm^3 [38,39]. Pure oxygen production, however, incurs high cost which makes it less competitive. Steam gasification is comparatively economical and brings high hydrogen yield [32] which is attributed to the utilization of the hydrogen content of steam in the reforming and shift reactions [40]. The thermal efficiency of conventional gasification i.e., moisture content less than 10–15% [41] decreases considerably as the biomass moisture content increases [42]. Therefore, for biomass with high moisture content of as high as 90–95%, supercritical water gasification is found to be a promising technology for hydrogen production [43–45]. Gil et al. [32] conducted experimental study to evaluate effect of different gasifying agents i.e. air, pure steam and steam–O_2 mixtures, on product gas gasification. Results from the study showed that steam gasification enhances the concentration of hydrogen in the product gas.

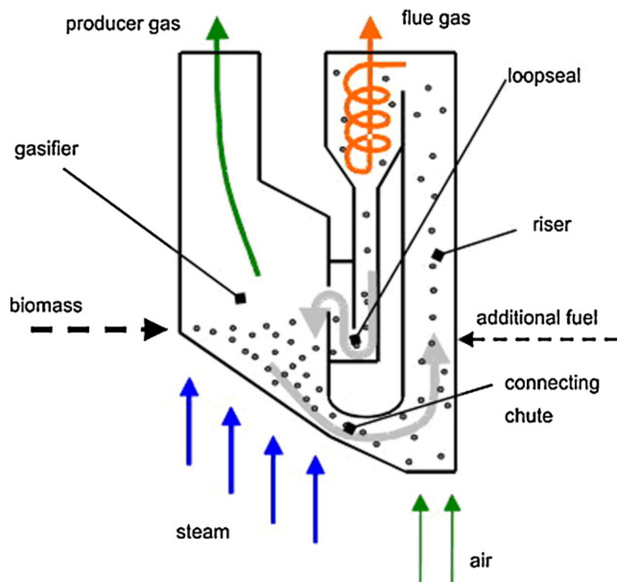

Fig. 1. Typical schematic drawing of a dual fluidized bed gasification reactor.
Source: Adapted from [149].

2.3. Types of biomass gasifier

Three types of gasifier are used for biomass gasification purposes i.e., fluidized bed-, fixed bed- and entrained flow gasifiers. Fluidized bed gasifiers have some advantages over fixed bed gasifiers including the uniform and controllable temperature distribution in the gasification zone and their ability to accommodate flexible feed rate and composition [46]. Such features make fluidized bed gasifiers attractive for large scale biomass gasification [47]. The depth analysis of the advantages and disadvantages of the three types of gasifier is reviewed by McKendry [41], and in a separate study Warnecke [48] reviewed the pros and cons of fixed and fluidized bed gasifiers. Nevertheless, regardless of gasifiers' configuration, biomass gasification is believed to occur in sequential steps of drying (100–200 °C), devolatilization (200–500 °C) and gasification (500–900 °C) [29].

Bubbling-, circulating- and twin-bed gasifiers are the three different types of fluidized bed gasifier. They differ in respect of fluidizing velocity and gas path [41]. Twin-bed gasifiers (Fig. 1) are of recent development in which the gasifier vessel is divided into two distinct fluidization chambers operated at two different gas velocities [37]. The design aimed to avoid mixing of gasification products with those from the combustion in order to obtain high purity hydrogen [49].

The type of transport of fluid inside gasifier differentiates one type of fixed bed gasifier from another. While biomass is often added from the top of the gasifier and flows downwards, depending on the flow direction of gasifying agents fixed bed gasifier can be updraft (counter-current), downdraft (co-current) and crossdraft (cross-current) [50]. In entrained flow gasifier the biomass and the gasifying agent move co-currently and the reactions occur in a dense cloud of very fine particles at high pressures (19.7–69.1 atm) and very high temperatures (i.e. >1000 °C) [51].

Due to the endothermic nature of biomass gasification, it necessitates external energy input. According to the ways to provide the required energy, gasifiers can further be grouped into autothermal and allothermal types. For autothermal gasifiers, partial oxidation of the biomass in the gasifier itself provides the necessary heat; air and oxygen gasification often employ such an approach [52]. Unfortunately, the oxidation generates exhaust gas which dilutes the product gas and reduces its heating value [49,52]. In contrast, allothermal gasifiers get the energy from external sources often in a form of bed materials carrying heat [53]; steam and CO_2 gasification are examples of this type. The combustion of the residual char outside the gasifier is also another method in which allothermal gasifiers can acquire the required energy [54]. To date, various research efforts have continued to integrate other forms of renewable energy with biomass gasification for the same purpose including direct sun energy [55,56], geothermal energy [57] and nuclear heat [49]. The main advantage of allothermal gasification systems is their ability to produce significantly higher H_2/CO ratios.

Needless to say, a desired target quantity and quality of gasification product gas should not compromise on energy consumption for production. Hence, the engineering-based techniques including modeling, simulation and heat integration are invaluable tools which can be used to optimize the trade-off taking into account the process, energy, economic and environmental constraints.

3. Development of mathematical models

Particularly with regards to biomass conversion into hydrogen via gasification, there exists a wide range of investigation relating to technologies and operating conditions, type of reactor and separation and purification equipment, type of biomass, and even type of fuels. Due to the inherent complexity of biomass gasification processes, modeling for simulation and prediction of performance of the processes is still an incipient activity [58].

In order to optimize the design and operation of biomass gasifier, an extensive investigation of the plant behavior depending on various operating parameter is required [59]. Often conducting experiments at large scales is problematic, undesirable in some cases for safety reasons and expensive as well [60]. Instead, as mathematical modeling gives a good representation of the chemical and physical phenomena occurring in the gasifier the resulting models can be used to study the plant behavior for use in optimizing the gasifier design and its operation (start up, shutdown, etc.) with minimal temporal and financial costs [29,59,60].

Warnatz et al. [61] stated that when considering a chemically reacting flow a system at each point in space and time is characterized by the specification of properties including pressure, temperature, velocity of flow, density and concentration of each species. Any change in the properties is the result of chemical reaction, fluid flow, molecular transport and radiation [61]. Hence, when modeling biomass gasifier depending on the required level of understanding on the system, one may take into account the influence of all or some of the above mentioned mechanisms i.e. chemical reaction, fluid flow, molecular transport and radiation. Upon completion of model development, the accuracy of the model needs to be verified and validated against the reality of the system, other than compared to other model's performance.The overall system efficiency and practicality of produced hydrogen for end-use also depends highly on efficiency of processes downstream the gasifier. However, to date—to our knowledge—modeling efforts in literatures have focused mainly on biomass gasifier alone. Hence, the reported mathematical models for biomass gasifier which are specifically used for hydrogen production can be classified into thermodynamic equilibrium models, kinetics models and neural network models [62]. Each type of models has their own strengths and limitations as discussed in the following sections.

3.1. Equilibrium models

The concept of chemical reaction equilibrium is based on the second law of thermodynamics as applied to reacting systems [63]. It is a state where species of a reaction system will no longer experience net change in concentrations over time. Hence, by employing

Table 1
Fundamental equations of equilibrium models [80].

Overall mass balance: (1) $F_{in}\sum_{in} n_{k,i}x_i = F_{out}\sum_{out} n_{k,i}x_i$ where $n_{k,i}$ is the number of atoms k of a molecule i, and x_i is the molar fraction of a component i.	Equilibrium constant of reaction j: (3) $K_j = \prod_i \left(\frac{P_i}{P_o}\right)^{v_{i,j}}$ which is related to temperature by: (4) $-RT\ln K_j = \Delta G^{\circ}{}_j$ where $\Delta G^{\circ}{}_j$ is the variation of standard Gibbs free energy of reaction j as function of temperature.
Overall energy balance: (2) $F_{in}\sum_{in} x_i H_i(T_{in}, P) = F_{out}\sum_{out} x_i H_i(T_{out}, P)$	(5) $G_{total} = \sum_{i=1}^{N} n_i \Delta G^{\circ}{}_{f,i} + \sum_{i=1}^{N} n_i RT \ln\left(\frac{n_i}{\sum n_i}\right)$

the governing equations describing the behavior of such state one can formulate an equilibrium model. Equilibrium models are important to predict the highest gasification or thermal efficiency that can be possibly attained for a given feedstock [64] and such a knowledge is vital for gasifier design and for selection of materials of construction [65,66].

On the whole, equilibrium models are suitable as a simulation tool for processes either whose duration is usually quite long with respect to the reaction time scale [62] or with gasification temperature higher than 800 °C [67]. The models possess generic applicability for simulating different gasifier configurations as they are independent of the gasifier design and not limited to a specified range of operating conditions [38,66,68]. There are two equilibrium modeling approaches widely used for predicting equilibrium composition of the product gas through minimization of the value of the Gibbs free energy, and those are stoichiometric and non-stoichiometric [69,70].

Stoichiometric models are based on equilibrium constants of independent set of reactions which can be associated with Gibbs free energy change as given by (Eq. (4)) [71]. Examples of models established on this approach include the work by [64,71–74]. On the other hand, the non-stoichiometric equilibrium modeling approach often referred as "*Gibbs free energy minimization approach*" is founded on the direct minimization of the Gibbs free energy of reaction species as defined in (Eq. (5)). This approach is common among many researchers [62,63,67,70,75–78] and it is claimed to be applicable for even more complicated reaction networks avoiding the need to identify the independent set of reactions. Table 1 includes the fundamental equations necessary to formulate an equilibrium model by employing one of the mentioned modeling approaches. The necessary coefficients for calculating thermodynamic properties of individual reaction species can be found from [79]. In the end, the solutions to the resulting equilibrium model equations can be obtained using different mathematical programming algorithms such as a built-in solver in MATLAB, fsolve. However, irrespective of the differences in the two approaches, both reported similar results [80].

Most of the reported thermodynamic gasification models focused on char reduction reactions assuming drying, pyrolysis and oxidation processes lumped together in a single reaction zone [81]. However, Ratnadhariya and Channiwala [66] recently reported a three-zone equilibrium model of biomass gasifier. The first zone is drying and pyrolysis, the second zone is oxidation and the third being the reduction zone. Such a model has an improved predictive capability since it more closely approximates the underling reality.

Florin and Harris [20] developed an equilibrium model for hydrogen production from steam gasification of eucalyptus coupled with in situ CO_2 capture using CaO as an adsorbent. The underlying idea behind the CO_2 capture is to alter the equilibrium composition of the product gas and subsequently promoting hydrogen production. The authors reported an increase in hydrogen concentration by 50–70 vol%. A similar conclusion was drawn by Guan et al. [82] utilizing a thermodynamic equilibrium calculation software named FactSage 5.2. Weil [83] conducted chemical equilibrium calculations which enhances the same concept by integrating gasification into cement manufacturing plant. The hot cement meal is used as a heat carrier and interestingly the CaO present in it operates simultaneously as an effective in situ CO_2-sorbent as well. In addition, such integration helps to minimize the cement manufacturing costs due to the marketing of hydrogen.

Although it is common to neglect tar while modeling gasification processes [29], Brown et al. [84] expanded the predictive capability of the chemical equilibrium calculations further for explaining the non-equilibrium behavior of gas, char and tar. The study incorporated the application of artificial neural networks into the modeling procedures to relate the changes in temperature differences to fuel composition and operational variables. Besides, equilibrium models coupled with exergy analysis are utilized to assess the practicality of biomass conversion into hydrogen via gasification [85–88].

Generally, equilibrium models are relatively easy to implement with faster convergence [81]. The models predict the highest attainable amount of hydrogen from gasification of a given biomass once the system reaches its equilibrium state. However, when developing equilibrium models it is often assumed that the system is at steady-state. Thus, in order to estimate the composition of the gasification product gas and the influence of various operating parameters at any point of space and time of the system, it is essential to develop a detailed kinetics model [60].

3.2. Kinetics models

Kinetics models take into account the reaction kinetics of the main reactions and the transfer phenomena among phases in biomass gasifier [65,81]. Such a model is capable for estimating the composition of product gas with varying operating conditions, which is an essential knowledge for designing, evaluating and improvement of gasifiers [65,81]. Often the hydrodynamics of the gasifier is also incorporated into kinetics modeling to assess the transport of gases from and to the reaction site.Rate laws (Eq. (6)) and kinetic parameters (Eq. (7)) make up the important components of a kinetics model [89]. The known Arrhenius Eq. (7) has a vital role in demonstrating the temperature dependence of the reaction rate. Upon obtaining these equations, if the model has not yet been able to be used to answer the model objective, conservation principles of the system, i.e., mass balances, energy balances, momentum balances and mole balance on each element should be incorporated.

$$r_i = k_i C_A^n C_B^m - k_{i,2} C_P^q \tag{6}$$

where r_i is the rate of i-th reaction, k is the Arrhenius constant for i-th reaction, C_i is the concentration of the component i with i is reactants A and B and product P, and n, m and q are orders of the reaction with respect to the component concentration.

$$k = A \times \exp\left(\frac{-E_a}{RT}\right) \tag{7}$$

where k is the reaction rate constant, A is the pre-exponential factor, E_a is the activation energy, R is the universal gas constant and T is the reaction temperature in Kelvin.

There are a number of softwares applicable platforms for modeling works including: spreadsheets for quick and easy arithmetic calculations and numerical mathematical solvers such as Mathematica and the ordinary differential equation (ODE) toolbox in MATLAB. It is worth to notice that suitable modeling software must be chosen based on its strength and limitations.

Based on the desired model prediction accuracy, when modeling biomass gasifier on reactor scale, the gasifier can be modeled

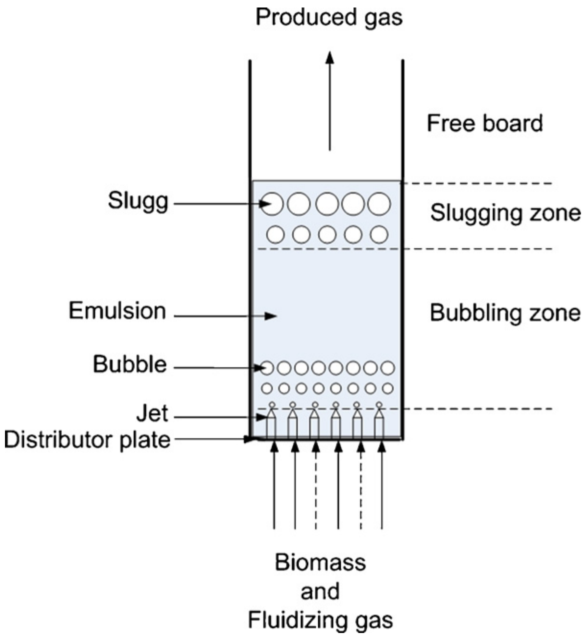

Fig. 2. Fluidized bed gasifier zones.
Source: Adapted from [92].

as plug flow reactor, stirred tank reactor and so on. Inayat et al. [90] constructed a kinetic model for biomass steam gasification for hydrogen production with in situ CO_2 capture considering the gasifier as a batch reactor. The work reported profiles on product gas composition that matches empirical results to some extent. Overall, in this paper the kinetics models for biomass gasifiers which are reported in literatures are reviewed being classified according to the gasifier types considered in the respective studies and presented as follows.On the whole, several mathematical models have been proposed for biomass gasification kinetics with different degrees of compliance, accuracy and adaptability to the different reactor configuration [91]. Nevertheless, one limitation of kinetics models stand out i.e., the models contain parameters that restricts their applicability to different process plants [38].

3.2.1. Fluidized bed gasifiers

Fluidized bed gasifiers are attractive technology for large scale production of hydrogen from biomass gasification. The kinetics models for fluidized bed gasifiers can be divided into three main groups, characterized by the number of phases accounted in the reactor: single-, double- and three-phase model [91]. Fig. 2 depicts the fluidized bed gasifier zones [92]. However, not all the indicated zones are considered by researchers during modeling development for various reasons including for model simplification purpose.

Fluidization is a complex phenomenon, and hence several authors reported kinetic models for gasifiers with various degrees of accuracy and details. Various assumptions are also included which aimed at either simplifying models for practical applications or due to limited experimental data in different areas. For instance, some assume drying and devolatilization are instantaneous phenomena [91], while others consider drying and devolatilization crucial components of the model [29,59]. Moreover, it is often assumed the gasification product gas to be free from tar [91] and some include a sub-model for tar generation and cracking phenomena [59].

An early work by Raman et al. [93] described a mathematical model for feedlot manure gasification kinetics in a fluidized bed gasifier. The model is reported to be the first of its kind to attempt to include the hydrodynamics of the gasifier and ultimately used to predict composition of gases produced. The model incorporated kinetic parameters for steam gasification of taken from literature. Various simplifying assumption were made in the course of the model development such as, the water-gas shift reaction was the only gas-phase reaction considered. However, the comparison of results from the model with experimental data suggested that cracking and reforming reactions involving the volatiles produced during devolatilization should be included in the model. Fiaschi and Michelini [91] developed a kinetics model for a fluidized bed gasifier and came to a conclusion that when comparing the effect of mass transfer and surface reaction kinetics on the whole gasification mechanism, the first prevails at the start of the process due to the high temperature level and successively, when the temperature is stabilized the latter plays a dominant role.

The summary of kinetics modeling works reported in literatures for fluidized bed biomass gasifier is included in Table 2. Despite some differences in bubbling and circulating types of fluidized bed gasifier [94], models developed for the bubbling process could be adapted to simulating circulating ones as long that new strategy to compute the freeboard or lean region were revised [94]. Two other exclusive literature reviews on modeling of fluidized bed biomass gasifiers can be found elsewhere [39], [95].

3.2.2. Fixed bed gasifiers

Nevertheless, fluidized bed gasifiers are not economical for small scale application [29]. Instead, for plants with low capacity fixed bed has no investment disadvantages [48]. Downdraft fixed bed gasifiers are the most widely used type of fixed bed gasifier. In case of updraft fixed bed gasifiers, since the tar produced during devolatilization stage does not go through the combustion zone, the tar content of the product gas from the gasifier is high. Hence, updraft gasifiers are less commonly employed type due to environment related concerns. The summary of kinetics modeling works reported in literature for fixed bed gasifiers is included in Table 3.

3.2.3. Entrained flow gasifiers

The need to provide very fine biomass feed i.e., particle size less than 0.1–0.4 mm and the high operating condition requirements deters wide use of entrained flow gasifiers [41]. Consequently, overall a few mathematical modeling works are available for entrained flow biomass gasifiers [96,97].

3.3. Artificial neural networks models

Artificial neural networks is a mathematical modeling approach which makes use of regression to correlate input and output streams to and from process units. It principally relies on large number of experimental data. While neural networks modeling takes a significant place in various other application areas, lately it has also received attention as a tool in renewable energy system prediction and modeling [98]. It demands less knowledge of the system phenomena as compared with equilibrium and kinetics modeling approaches.

Neural networks models are reported to improve accuracy of composition prediction of gasification product gas [62]. However, the neural networks modeling require intensive experimental data and such data from biomass gasification processes are not readily available in literatures. For this reason, not many works on neural networks model development are reported. Kalogirou [98] recently reviewed the literatures on artificial neural networks in renewable energy systems applications. Guo et al. [99] developed an artificial neural network model which was used to simulate biomass gasification processes and emphasized the applicability and effectiveness of the models.

Table 2
Summary of the kinetics modeling studies on fluidized bed biomass gasifiers.

Reference	Reactor type and its operation mode, biomass & gasifying agent studied	Model dimension and multiphase modeling approaches	Fundamentals of the model formulation
Fiaschi and Michelini [91]	BFBG Dynamic Sawdust Air and oxygen	1 D Two phase (bubble and dense)	Reaction kinetics (in dense phase), mass transfer between the two phases, quantitative estimation of local bubble and particle properties Freeboard area considered chemically inert Drying and devolatilization considered instantaneous Tar content of the product gas is assumed to be assimilated in CH_4
Hamel and Krumm [59]	BFBG Straw Air, air-stream and oxygen- steam	Two phase (bubble & emulsion phase)	Reaction kinetics (drying and devolatilization), bed and freeboard hydrodynamics Predict amount of tar in product gas
Sadaka et al. [92]	BFBG Dynamic and steady state Agricultural waste- Straw Air-steam	Two phase (bubble & emulsion)	Kinetics parameters and hydrodynamics, transport and thermodynamic properties of fluidized bed Free energy minimization technique is used to calculate gas mole fractions at the devolatilization stage
Lu et al. [150]	BFBG Steady state Wood	1 D Two phase (bubble and emulsion)	Reaction kinetics, axial gas dispersion in the two phases Pyrolysis (devolatilization) is taken to be instantaneous
Kaushal et al. [29]	BFBG Steady state Wood Air, oxygen, steam and mix of these gases	1 D Two phase (bubble and emulsion)	Global reaction kinetics, mass & energy balances, hydrodynamics (for bubbling regime of fluidization), material properties and sub-model for tar generation and cracking
Gordillo and Belghit [49]	BFBG Dynamic and steady state Biomass char Steam	1 D Two phase (bubble and emulsion)	Reaction kinetics, mass and energy balances Mass and heat transfer occurs only between the bubbles and emulsion and the emulsion and the solid phases
Radmanesh et al. [151]	BFBG Dynamic Beech wood Air	1 D Two phase (bubble and emulsion)	Takes into account pyrolysis and various heterogeneous and homogeneous reaction kinetics as well as the hydrodynamics of the bed and the freeboard. Solid phase (char) is modeled by the countercurrent back-mixing (CCBM) model. The study used a first-order kinetic model proposed by Borosan et al. [98] and a kinetic model by Bryden and Ragland [99] to represent the homogenous cracking of tar and combustion of tar respectively.
Corella and Sanz [152]	CFBG Steady state Pine wood Air	1 D Core-annulus approach	Reaction kinetics, mass and energy balances and a sub-model for the tar generation-elimination and some empirical aspects
Petersen and Werther [36]	CFBG Dynamic Sewage sludge Air and CO_2/N_2-mixture	A 1.5-dimensional (pseudo two dimensional) Core-annulus approach	Fluid dynamics of a CFBG, kinetics of pyrolysis and gasification steps Benzene taken as model component for biomass tar
Petersen and Werther [153]	CFBG Dynamic Sewage sludge Air	3 D Core-annulus approach	Uses continuous radial profiles of velocities and solids hold-up with regard to the description of fluid mechanics
Kaushal et al. [154]	TBFBG Steady state Biomass char Steam and air	1 D Bottom bed: two phase (bubble and emulsion phase) and Freeboard: core-annulus approach	Sub-models for bed hydrodynamics, conversion and conservation (mass and energy)

4. Development of simulation models

Upon the development of the mathematical model for a given biomass processing system and its validation, the model can be utilized to predict or simulate the behavior and/or criteria and/or performance of the system. When it comes to simulating a complete flowsheet, due to its complexity involving hundreds of equations and variables, it is often advantageous to use process simulators to evaluate the process performance depending on different operating conditions in reasonable time with minimal effort. Built-in mathematical models being the building-blocks of process simulators, here users are instead expected to develop the process flowsheet and provide the required input data.

Process simulation can be of at steady state where the variation of process properties over time is ignored and dynamic simulation which accounts the time dependence of processes by incorporating the accumulation terms in the conservation equations i.e., mass, energy and momentum balances. The latter has advantages as it depicts the behavior of processes in real time change which is essential for process control activities. Steady state simulators which can be used in simulating biomass gasification for hydrogen production include ASPEN PLUS, PETRONAS iCON, IPSEpro, HYSYS,

Table 3
Kinetic models for fixed bed biomass gasifiers.

Reference	Reactor type, mode of operation, biomass studied and gasifying agent	Fundamentals of the model formulation	Parameter studied
Chen and Gunkel [155]	Downdraft Wood Air	Non isothermal particle model applying principles of thermodynamics, transport processes, hydrodynamics of solid and gas flows, and the mass and energy balances. One dimensional	Feedstock moisture contents, particle sizes, reactor insulation, input air temperatures and gasifier loads, dimension of gasification zone
Blasi [156]	Downdraft Dynamic Air	Heat and mass transfer across the bed, reaction kinetics (drying, pyrolysis, char combustion and gasification, gas phase combustion and thermal tar cracking).	Predictions of the gas composition and the axial temperature profile
Giltrap et al. [157]	Downdraft Steady state Air	Reaction kinetics (reduction zone)	Output gas composition
Babu and Sheth [158]	Downdraft Steady state Biomass char Air	Reaction kinetics (reduction zone)	Char reactivity factor Composition and temperature profiles for the reduction zone
Gobel et al. [159]	Downdraft Dynamic Wood Air and steam	Mass and energy balances in simply one dimensional flow reaction kinetics in the gas phase and Langmuir–Hinshelwood correlation describing reaction kinetics in the char The char consisted pure carbon Considers no tar in the gasification zone	Char reactivity, bed height temperature profile, gas composition and its heating value
Tinaut et al. [160]	Downdraft Steady state Any type of biomass Air	Mass and energy conservation equations One dimensional	Biomass particle mean diameter, air flow velocity, gasifier geometry, composition and inlet temperature of the gasifying agent and biomass type
Sommariva et al. [161]	Updraft Steady state Refuse derived fuels (RDF) and wastes Air	Heat and mass transport resistances and chemical kinetics (both at the reactor and particle scale)	Particle size Temperature Feed flow rate

SuperPro Designer, Pro/II, ChemCad, etc. For dynamic simulation, MATLAB, ANSYS Fluent, ANSYS CFX, CFD2000 are good examples of software.

4.1. Process simulation models

ASPEN is a standard process flowsheet simulation tool which can be used to simulate the biomass gasification process [100,101]. Several ASPEN PLUS simulation models have been reported in the literatures which investigates the effect of different operating variables on the performance of the process. Shen et al. [54] simulated air-steam gasification of biomass in interconnected fluidized bed gasifier. A steady state simulation model for the process is constructed in the ASPEN PLUS simulator. The model assumed chemical equilibrium is reached for the reactions and it was used to study effect of temperature and S/B ratio on hydrogen yield.

Doherty et al. [102] reported an equilibrium simulation model constructed in ASPEN PLUS simulator. The model allowed for predicting syngas composition, heating values and conversion efficiencies while evaluating the influence of different variables including gasification temperature, equivalence ratio and level of air preheating on gas composition. The results from the study showed that air preheating enhanced H_2 and CO production and it is more effective at low equivalence ratios and should not be used for ERs greater than 0.35. Nikoo and Mahinpey [103] presented an ASPEN PLUS simulation model which is based on kinetics of a gasification process in an atmospheric fluidized bed gasifier. The model takes into account both the hydrodynamic parameters and reaction kinetics of the process. The model is used to observe the effect of reaction temperature, equivalence ratio, S/B ratio and particle size on hydrogen production.

Tan and Zhong [104] reported an equilibrium simulation model for steam gasification of sawdust which is implemented in ASPEN PLUS simulator. The model is used to investigate effect of process parameters including gasification temperature, pressure and steam to biomass ratios (S/B). The results showed hydrogen content of up to 60 vol%. Moreover, it was observed that higher S/B ratios favored hydrogen yield while increasing temperature above 700 °C is observed to have adverse effect on hydrogen yield.

Pröll et al. [105] simulated gasification of biomass in dual fluidized bed gasifier for enriched H_2 gas production in equation-oriented steady state simulation software named IPSEpro. The model was used to investigate a case where CaO was used for selective transport of CO_2. This work observed that the CO_2 transport concept not only enhances the hydrogen content in the product gas but also makes the process more energy efficient. Schuster et al. [38] also reported a simulation model of a decentralized combined heat and power station based on a dual fluidized bed steam gasifier using IPSEpro process simulator. The net electric efficiency of the system is reported to be about 20%. Among many parameters evaluated, the results of the sensitivity analysis showed that gasification temperature and fuel oxygen content are the most significant parameters which determine the chemical efficiency of the gasification.

Detournay et al. [106] developed a simulation model using HSC Chemistry 5.1 software which is based on Gibbs Energy MINImization (GEMINI code). Ahmad et al. [107] reported an equilibrium simulation model for hydrogen production via oxygen assisted gasification of empty oil palm fruit bunch (EFB) with in situ adsorption of CO_2. The simulation model is constructed in PETRONAS iCON and used to evaluate the influence of temperature (600–1000 °C) and S/B ratio (0.1–1) on hydrogen yield and product gas composition. Ahmad et al. [108] also simulated flowsheet of pressurized gasification process of biomass coupled with CO_2 adsorption for hydrogen production using a PETRONAS iCON. The effect of parameters such as pressure, temperature and S/B ratio on the hydrogen yield was investigated. Hydrogen yield was predicted to be increasing with pressure, temperature and S/B ratio in this high pressure gasification system.

Inayat et al. [109] analyzed the influence of the temperature, S/B and sorbent/biomass ratios on steam gasification flowsheet using MATLAB. They observed that the thermodynamic efficiency of the gasifier increased with temperature and S/B ratio. The maximum efficiency obtained about 84% at S/B ratio of 2.0. They predicted that by using CaO as a sorbent in the gasifier, the efficiency can be increased by 10% compared to the conventional gasification method. Inayat et al. [110] also developed process simulation model which was used as a platform to investigate the effects of process parameters on the production of hydrogen rich gas from EFB using a single-pass fluidized bed gasifier. Based on the results, the maximum hydrogen purity predicted is 71 mol% at 1150 K at outlet of the gasifier unit with the yield of 107.3 g/kg of EFB. The mass conversion efficiency increases with increasing temperature and is found to be at the maximum at 84.7% at 1100 K. Ahmad et al. [111] used a simulation approach to study the influence of temperature, S/B and sorbent/biomass ratios against the thermodynamic efficiency for hydrogen production from steam gasification taking biomass as pure char. The results show that the thermodynamic efficiency depends on feed stock quality and vary with the operating conditions. The model predicts an increment in the thermodynamic efficiency from 66.5% to 83.3% within the temperature range of 900–1100 K.

4.2. Computational fluid dynamics simulation models

Computational fluid dynamics (CFD) is the science of predicting fluid flow, heat transfer, chemical reaction and other related phenomena by solving numerical set of the governing mathematical equations which are mostly based on conservation equations i.e., mass, heat and momentum. However, due to the complexity of the gasification process i.e., involving many phases and various chemical and physical interactions among them not much works are available concerning development of mathematical CFD model to be used for simulation purposes. Generally, results of CFD analysis are relevant for conceptual studies of new design, detail product development, troubleshooting and redesign. Besides, CFD modeling also is cost saving, timely, safe and easy to scale-up [112].

Various numerical techniques have been employed in the solution of the CFD model equation and the most widely use numerical technique is discretization method including finite difference, finite element and finite volumes method. Finite volume is now the most commonly used approach in CFD code for its ease in the understanding, programming and versatility. The most routinely used commercial codes include ANSYS Fluent, ANSYS CFX, CFD2000 and many others [112]. Most reported CFD simulation models in literatures are either for coal gasification or for biomass combustion/gasification in entrained flow gasifiers [113–115]. Recently, Rashidi [116] has discussed CFD simulation of biomass gasification using detailed chemistry. Papadikis et al. [117,118] and Xue et al. [119] used CFD modeling approach for simulating fast pyrolysis of biomass in fluidized bed gasifiers. In addition, Pepiot et al. [120] reported a CFD model for both biomass gasification and pyrolysis. Generally, for better result comparisons and to improve the modeling and simulation of biomass gasification in CFD, it is necessary to develop more CFD models. Thus, the development of CFD models needs further research and development effort.

5. Process optimization

In order cope with the current energy and environment crisis, industries need to convert operations of a plant/system into an efficient system in terms of utilization of energy, resources and materials. Generally, there are two systematic mathematical approaches to make a system efficient: one is to perform heat integration studies to maximize heat recovery in the system and hence reducing the utility requirement; and two is to find optimal operating conditions to minimize the cost of the production/operation. The heat integration approach will be discussed in Section 6 of this paper.

Optimization can be applied to search for the optimal conditions and/or geometric parameters that give extremes of selected objective functions in the frame of previously defined constraints [121]. For instance, a mathematical optimization model can be developed to find the minimum production cost of a product subject to the performance and constraints of a selected processing system. The objective of an optimization problem can also be the maximum product yield or purity, maximum profit, minimum production time, best combination of raw materials that generate maximum profit, best process configuration that yields the maximum operating efficiency, the lowest carbon emission while fulfilling the demand, etc. An optimization problem must have the degrees of freedom of more than zero, i.e., there must at least one free independent variable in the calculation that can be changed by the optimizer/solver to achieve minimum or maximum objective function making it different from simulation where the degree of freedom is zero.

Among the tools available for optimization are General Algebraic Modeling System (GAMS), MATLAB Optimization Toolbox, Solver in Microsoft Excel, LINDO, SolvOpt and many more. Takriff et al. [122] conducted literature review on integrating optimization modules into chemical process simulation softwares. The authors identified the use of readily available process simulation software as an interface to structural and continuous optimization as possible and promising strategy.

Kamarudin et al. [123] conducted mathematical optimization to investigate hydrogen demand and determine the optimum hydrogen delivery network employing truck transportation in Malaysia. The overall objective function the authors used to minimize the total investment of production plant is given by (Eq. (8)). The objective function is optimized while being subjected to a combination of various constraints. Hugo et al. [124] presented a generic optimization-based model for the strategic long-range investment planning and design of future hydrogen supply chains using Mixed Integer Linear Programming (MILP) techniques.

$$Min(x)\left[\left[\sum_{g_i}\left(\sum_{p_i}1.3FC\right)\right] + \sum_{g_n}\left(\sum_{T_m}[CC+NV(VC)]+\sum_{S_o}SC\right)\right]\alpha\beta \tag{8}$$

As the number of literatures directly dealing with optimization of biomass gasification for hydrogen production is limited, it is our belief that reviews on biomass processing evolving gasification will

be beneficial for future development of optimization models on the area. Gasification is one route which can be used for producing ethanol from biomass. Employing an optimization algorithm previously developed for the design of integrated process water network [125], Cucek et al. [126] reported an optimization work conducted on integrating energy, water and process technologies for simultaneous production of ethanol and food from corn plant. Ahmetovic et al. [127] also reported energy optimization model for the design of corn-based ethanol plants. Verda and Nicolin [128] performed a thermo-economic optimization of a molten carbonate fuel cell (MCFC) stack integrated with a micro gas turbine for electricity generation coupled with a pressure swing absorption system (PSA) for hydrogen production. Muis et al. [129] developed optimization model used for analysis of biomass usage for electricity production with the main aim of reducing CO_2 in Malaysia.

6. Heat integration

Heat integration maximizes savings by making processes energy efficient and self sustainable through utilizing recovered process heat which significantly reduces the process dependency on external energy supply [130]. Process to process integration and integration of heat engines and heat pumps into processes are the known methods for heat integration. Heat integration approach using pinch analysis [131] has been accepted by many major companies to revamp their plants to reduce their energy consumption. This way, less fuel (hence operating cost) is required to generate energy in the form of steam and electricity to support their operations. Tools available for heat integration studies are such as Microsoft Excel and SPRINT by Centre for Process Integration, University of Manchester.

Pavlas et al. [132] used heat integration methodology and effectively integrated heat pump with biomass gasification. The authors reported the availability of large amount of waste heat from biomass gasification and proved the feasibility of the integration to allow efficient utilization of the available heat for heating and cooling requirements. Sadhukhan et al. [100] performed heat integration analysis on biomass integrated gasification combined cycle processes. They modeled a combined heat and power generation plant from gasification of low-cost, fourth generation biomass waste feedstocks in ASPEN simulator and performed a heat integration study to demonstrate the potential via an energy production cost analysis that included detailed discounted cash flow profile. Heyne et al. [133] reported a study on synthetic natural gas production from biomass gasification using integrated process design in ASPEN PLUS. The authors applied pinch analysis for the optimal internal heat recovery calculation within the process. Smejkal et al. [134] presented a model for energy production from biomass gasification using process integration approach. The model predicted high efficiency and low operating cost of process.

Inayat et al. [135] performed heat integration analysis for a process flowsheet developed for hydrogen production from steam gasification of EFB with in situ CO_2 capture using the SPRINT software. The authors employed pinch analysis to obtain an energy efficient and self-sustained system. Their result analysis showed that considerable saving can be obtained for steam production using heat integration application as there is a large amount of available waste heat from the gas cleaning and cooling units. Ahmad et al. [136] developed a process simulation model of a selected flowsheet for a biomass gasification plant for hydrogen production using ASPEN PLUS software. They performed heat integration study on the plant flowsheet using pinch analysis and was carried out in SPRINT, University of Manchester Process Integration Software. The minimum temperature difference was set to be 10 K. The results reported possible energy savings of approximately 72% in hot utilities and 88% in cold utilities via the heat integration analysis.

7. Cogeneration potential

Cogeneration also known as CHP is a power generation system which leads to an improvement in the overall system efficiency. While the different benefits of the cogeneration technology such as energy, economic and environmental benefits, are well acknowledged due to tight emission regulations direct combustion of biomass or fossil fuels for cogeneration applications are not attractive. Hence, the environment-friendly and biomass-derived hydrogen is expected to play a major role in substituting fossil fuels in CHP generation applications. Biomass fuelled cogeneration system is one of the key energy technologies of the future since it combines the merits of renewable energy sources and hydrogen energy systems [137]. Moreover, it is reported that the share of biomass in combined heat and power production is expected to increase in the future and decentralized combined heat and power plants are of interest to avoid the cost associated with biomass transportation [23]. Generally, cogeneration supports sustainable biomass-based hydrogen integration into existing large energy system [23] and promising work is being done on the area.

Available options for biomass based CHP systems in the output range of 1MWe include internal combustion engines, microgas turbines and fuel cells [21]. The integrated gasification combined cycle (IGCC) is an early application [138]. However, recently this trend has changed. In literatures biomass gasification for hydrogen production has been studied while integrated with power producing technologies such as evaporative gas turbines [139], micro-turbines [23,139,140], fuel cells [21,23,40,52,128,140–142] and gas engines [143].

Improved gas turbines are under development for use with H_2 and H_2-rich gases [144–146]. Fuel cells are emerging as an important component of a renewable energy future for many utility and mobile applications. A very promising technique is to obtain the required hydrogen from gasification of biomass. In a separate study, Rosen and Scott [147] investigated the cogeneration potential of several types of fuel cell system (phosphoric acid, alkaline, solid polymer electrolyte, molten carbonate and solid oxide) using energy and exergy analysis. Recently Colpan et al. [137] concluded that among the different types of fuel cells, MCFC and solid oxide fuel cells (SOFC) are considered as the most promising ones for hydrogen fuelled fuel cells due to their high operating temperature level, flexibility to different fuel and greater tolerance to contaminants. Biomass gasification processes are more commonly integrated to gas turbine based combined heat and power generation systems. However, efficiency can be greatly enhanced through the use of more advanced power generation technology such as SOFC [138].

Bang-Møller and Rokni [23] conducted a modeling study on three combined heat and power systems based on biomass gasification. In this system, the product gas from biomass gasification is converted in a micro gas turbine (MGT) in the first system, in a solid oxide fuel cell in the second system and in a combined SOFC–MGT arrangement in the third system. The authors reported that by combining the SOFC and MGT, the unconverted syngas from the SOFC is used in the MGT to produce more power and the SOFC is pressurized, which improves the efficiency to as much as $\eta_{el} = 50.3\%$. The optimal operating pressure ratio for the SOFC–MGT combination is also reported to be 2.5. Similarly, Abuadala and Dincer [40] studied an integrated process of steam biomass gasification and a SOFC and concluded that such an integration brings positive value to the overall system efficiency. For the integration of fuel cells into biomass gasification, cogeneration relevant performance

figures which could be investigated for different system configurations and cell parameters may include fuel utilization, fuel flow rate, operation voltage and extent of internal fuel reforming [148].

8. Conclusion

The purpose of this paper has been to review the various research works encompassing mathematical models, simulation models, heat integration, cogeneration and optimization which are contributing to the development of hydrogen as an energy carrier. From this review of literatures the following conclusions can be drawn:

- Biomass gasification is a sustainable and economical technology for producing hydrogen.
- Practicality and availability of hydrogen for end uses is highly dependent on several factors including technological, economic and social. Hence, to bring the future hydrogen-based economy into reality, issues on the respective areas should be addressed appropriately.
- From this review of literatures, it is learned that there exists limited data from large-scale biomass gasifiers for uses in mathematical model development and for model validation purposes. Hence, for wider and more reliable application of mathematical models for process optimization it is necessary to obtain data from large-scale operations of gasifiers.
- Each modeling approaches discussed has their own strengths and limitations. Mathematical models are essential for process optimization purposes to find optimal operating conditions which leads better process performance. However, the model development process could be tiresome. On the other hand, simulation models are easy for simulating a complete flowsheet avoiding the need to deal with large number of equations and variable. Nevertheless, simulation models could not be used for process optimization purposes. modeling effort employing the computational fluid dynamic modeling approach requires further research and development to enhance the understanding of biomass gasification processes.
- Integrating biomass gasification into power producing devices such as fuel cell and micro-turbines is an emerging and promising technology which allows the sustainable integration of the biomass-based hydrogen into the existing large energy systems.
- Evaluating the overall system performance in terms of energy usage, economic benefit and product distribution strategy using mathematical optimization strategies still require further research and development to assess effective implementation of hydrogen in energy and transportation systems.

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